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FORM PTO-159 (REV. 12-2001)		U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY'S DOCKET NUMBER MR2685-110
TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371				U.S. APPLICATION NO. (if known, see 37 CFR 1.5) 10/069617
INTERNATIONAL APPLICATION NO. PCT/KR00/01205	INTERNATIONAL FILING DATE 24 October 2000	PRIORITY DATE CLAIMED 6 September 2000		
TITLE OF INVENTION METHOD FOR PREPARATION OF CLAY-DISPERSED POLYMER NANOCOMPOSITE				
APPLICANT(S) FOR DO/EO/US Moon Bae Ko, et al.				
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:				
1. <input checked="" type="checkbox"/> This is a FIRST submission of items concerning a filing under 35 U.S.C. 371.				
2. <input type="checkbox"/> This is a SECOND or SUBSEQUENT submission of items concerning a filing under 35 U.S.C. 371.				
3. <input checked="" type="checkbox"/> This is an express request to begin national examination procedures (35 U.S.C. 371(f)). The submission must include items (5), (6), (9) and (21) indicated below.				
4. <input type="checkbox"/> The US has been elected by the expiration of 19 months from the priority date (Article 31).				
5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. 371(c)(2))				
a. <input checked="" type="checkbox"/> is attached hereto (required only if not communicated by the International Bureau).				
b. <input type="checkbox"/> has been communicated by the International Bureau.				
c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US).				
6. <input checked="" type="checkbox"/> An English language translation of the International Application as filed (35 U.S.C. 371(c)(2)).				
a. <input checked="" type="checkbox"/> is attached hereto.				
b. <input type="checkbox"/> has been previously submitted under 35 U.S.C. 154(d)(4).				
7. <input type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))				
a. <input type="checkbox"/> are attached hereto (required only if not communicated by the International Bureau).				
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d. <input type="checkbox"/> have not been made and will not be made.				
8. <input type="checkbox"/> An English language translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371 (c)(3)).				
9. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4))				
10. <input type="checkbox"/> An English language translation of the annexes of the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)).				
Items 11 to 20 below concern document(s) or information included:				
11. <input type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98.				
12. <input checked="" type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included				
13. <input type="checkbox"/> A FIRST preliminary amendment.				
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METHOD FOR PREPARATION OF CLAY-DISPERSED POLYMER
NANOCOMPOSITE

5 TECHNICAL FIELD

The present invention relates, in general, to a method for the preparation of a clay-dispersed polymer nanocomposite and, more particularly, to the use of an oxygen atom-carrying polymer with thermodynamical compatibility with a binder resin in preparing a clay-dispersed polymer nanocomposite, thus delaminating clay completely to silicate lamellas.

PRIOR ART

With the aim of overcoming limited physical properties of general-purpose plastics, the techniques for preparing clay-dispersed organic/inorganic composites are characterized in that clay ores of silicate layered structures are delaminated into silicate layers at nano levels and the silicate layers are dispersed in polymer resins, thereby upgrading general-purpose plastics to engineering plastics. Silicate layer, a structural unit of clay, is however difficult to disperse in polymer resins owing to its strong van der Waals attraction. To circumvent this problem, intercalants are suggested. According to a compounding technique, for example, an intercalant with a low molecular weight is introduced into the silicate layered structure to aid the penetration of polymer

resins into the structure, thereby delaminating clay ores and dispersing the silicate layers in the resins.

However, the conventional compounding technique has such a problem that, when nanocomposites are prepared from some resins, including poly(styrene-co-acrylonitrile)copolymer resins, poly(acrylonitrile-co-butadiene-co-styrene) copolymer resins, and poly(vinylchloride) resins, it is virtually impossible to completely delaminate clay ores to the extent of a silicate layer, which is a structural unit of clay. In the nanocomposites of such resins, prepared by the conventional compounding technique, in fact, there are observed blocks of nano scales, which result from the aggregation of several to tens silicate lamellas [see, *Korea Polymer Journal*, 7, 310 (1999); *ibid*, 8, 95 (2000); *ibid*, 8, 120 (2000)]. The reason is that a nitrogen and a chlorine atom, which are contained in the repeating unit acrylonitrile of poly(styrene-co-acrylonitrile) or poly(acrylonitrile-co-butadiene-co-styrene) copolymer resins and the repeating unit vinyl chloride of poly(vinylchloride) resin, respectively, is smaller in electronegativity than is an oxygen atom, which is present within organophilic clay, so that it is not easy for the polymer chain to penetrate between silicate layers.

DISCLOSURE OF THE INVENTION

Leading to the present invention, the intensive and thorough research on the preparation of clay-dispersed polymer nanocomposites, conducted by the present inventors, resulted in the finding that a polymer carrying oxygen atoms can itself penetrate into silicate structures and, when constituting a matrix for the composites, along with other polymers, help penetrate the binder resins into layered structure of silicates if they are thermodynamically compatible with the oxygen-carrying polymer.

Therefore, it is an object of the present invention to overcome the above problems encountered in prior arts and to provide a method for the preparation of clay-dispersed polymer nanocomposites, in which clay ores are completely delaminated to a silicate layer, which is a constituent minimum unit.

Based on the present invention, the object could be accomplished by a provision of a method for preparing a clay-dispersed polymer nanocomposite, which comprises introducing an oxygen-carrying polymer, alone or in combination with a thermodynamically compatible resin, as a matrix resin.

BRIEF DESCRIPTION OF THE DRAWINGS

Fig. 1a shows X-ray diffraction spectra of the nanocomposites of the present invention according to mixing periods of time.

Fig. 1b shows an X-ray spectrum of the clay-dispersed poly(styrene-co-acrylonitrile)copolymer nanocomposite according to the present invention, along with that of the clay-dispersed poly(ϵ -caprolactone) nanocomposite.

Fig. 2 shows X-ray diffraction spectra of the nanocomposites prepared at various temperatures according to the method of the present invention.

BEST MODES FOR CARRYING OUT THE INVENTION

The present invention pertains to a compounding technique of preparing a clay-dispersed polymer nanocomposite, in which a matrix resin, including a poly(styrene-co-acrylonitrile)copolymer, poly(acrylonitrile-co-butadiene-co-styrene) copolymer or poly(vinylchloride), is inserted into silicate structures with the aid of an intercalant, characterized in that a poly(ϵ -caprolactone) polymer is used.

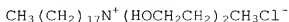
Poly(ϵ -caprolactone) carries oxygen atoms within the repeating units of its backbone, as well as being thermodynamically compatible with poly(styrene-co-acrylonitrile) copolymer, poly(acrylonitrile-co-butadiene-co-styrene) copolymer and poly(vinylchloride).

Useful in the present invention is a poly(ϵ -caprolactone) resin ranging, in molecular weight, from 10,000 to 100,000.

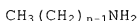
It is preferable that poly(ϵ -caprolactone) is used at an amount of 20-40 % by weight based on the total weight of the nanocomposite. For instance, if poly(ϵ -caprolactone) is added at an amount less than 20 % by weight, its compatibilization effect is too weak to bring about complete delamination into clay. On the other hand, more than 40 % by weight of poly(ϵ -caprolactone) may shield the expression of characteristic physical properties of other matrix resins.

In the present invention, clay to be dispersed in the matrix is treated with an intercalant in order to increase the dispersity of clay. The intercalant is preferably selected from the group consisting of hydroxyethylmethyloctadecylammonium chloride, which is represented by the following chemical formula 1; amines such as octyl amine, decyl amine, dodecyl amine, hexadecyl amine, and octadecyl amine, which are represented by the following chemical formula 2; and secondary amines represented by the following chemical formula 3:

Chemical Formula 1

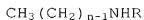


Chemical Formula 2



wherein n is 8, 10, 12, 16 or 18.

Chemical Formula 3



wherein n is an integer of 8-18 and R is a hydrocarbon.

As a constituent for clay, montmorillonite, which
5 swells in water, is preferably used.

Besides the polymer resins mentioned above, any
of the resins which are thermodynamically compatible
with poly(ϵ -caprolactone) can be used in the present
invention. chlorinated polyethylene is an example of
10 the thermodynamically compatible resins.

Preparation of clay-dispersed polymer
nanocomposites according to the present invention can
be achieved in either a one-pot or a two-pot manner.

As for the one-pot manner, it is conducted by
15 mixing a binder resin, poly(ϵ -caprolactone), and
organophilic clay and extruding them together. On the
other hand, according to the two-pot manner, poly(ϵ -
caprolactone) is mixed or extruded with an
organophilic clay to give a master batch, followed by
20 mixing with a binder resin for extrusion.

The following examples are set forth to illustrate
more clearly the principles and practice of this
invention to one skilled in the art. As such, they are
not intended to limit the invention, but are
25 illustrative of certain preferred embodiments.

EXAMPLE 1

Preparation of Poly(ϵ -caprolactone) Nanocomposite

An organophilic clay, such as that manufactured from Southern Clay Products Inc. U.S.A., identified as Closite 30A, was dry-blended at an amount of 5 % by weight with poly(ϵ -caprolactone) with a molecular weight of 80,000 at 100 °C at 120 rpm in a Minimax molder for various periods of time to obtain nanocomposites.

With reference to Fig. 1a, there are shown X-ray diffraction spectra of nanocomposites according to mixing periods of time. As seen in the spectra, the X-ray diffraction peak at around 2.5° disappears from the spectrum of the nanocomposite obtained when the mixing was continued for 20 min or longer; thus the silicate was completely delaminated with the aid of the intercalant in the nanocomposite.

EXAMPLE 2

Preparation of Clay-Dispersed Poly(styrene-co-acrylonitrile)copolymer Nanocomposite Using Clay-Dispersed Poly(ϵ -caprolactone) Nanocomposite

The clay-dispersed poly(ϵ -caprolactone) nanocomposite prepared in Example 1 was mixed at a weight ratio of 2:3 with a poly(styrene-co-acrylonitrile)copolymer resin, after which blending of the mixture was carried out at 100 °C for 5 min in a Minimax molder at 120 rpm to afford a clay-dispersed poly(styrene-co-acrylonitrile)copolymer nanocomposite.

EXAMPLE 5

Preparation of organophilic clay from Octadecylamine
and Preparation of Clay-Dispersed Poly(styrene-co-
acrylonitrile)copolymer Nanocomposite Using the Same

5 In a 3,000 ml beaker were charged 1,500 ml of
deionized water, 8.1 g of octadecylamine and 3.0 ml of
hydrochloric acid and the solution was heated to 80 °C
with stirring (Solution A). 1,500 ml of deionized
water and 30 g of clay, such as that manufactured by
10 Kunimine Co. Japan, identified as Kunifia F, were added
into another 3,000 ml beaker and heated to 80 °C. This
clay dispersion was poured to the Solution A and
stirred for 1 hour, after which a white precipitate was
separated by filtration. After being washed with
15 distilled water maintained at 80 °C, the precipitate
was dried to give 32.1 g of organophilic clay. This
was ball-milled to obtain a powder (C18-MONT) with a
size of 50 μm . Based on data of X-ray diffraction
tests, the space between silicate lamellas was
20 calculated to be 19 Å. The nanocomposite was measured
to comprise 74.0 % by weight of the content of pure
clay as measured by thermo gravimetric analysis. Taken
together, the X-ray diffraction data and the thermo
gravimetric analysis data demonstrate that the clay has
25 been transformed to organophilic clay.

To a polymer matrix in which poly(ϵ -caprolactone)
with a molecular weight of 10,000 was blended at an
amount of 30 % by weight with a poly(styrene-co-

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acrylonitrile)copolymer, the above organophilic clay (C18-MONT) was added at an amount of 5 % by weight and dry-blended. The resulting blend was extruded at 160 °C at 250 rpm to give a clay-dispersed poly(styrene-co-acrylonitrile)copolymer nanocomposite in which the silicate of the organophilic clay was completely delaminated to silicate layers.

EXAMPLE 6

One Pot-Type Preparation of Clay-Dispersed Poly(acrylonitrile-co-butadiene-co-styrene) Copolymer Nanocomposite Using Twin-Screw Extruder

To a polymer matrix in which poly(ϵ -caprolactone) with a molecular weight of 10,000 was blended at an amount of 30 % by weight with a poly(acrylonitrile-co-butadiene-co-styrene) copolymer, organophilic clay, such as that manufactured by Southern Clay Products Inc. U.S.A., identified as Closite® 30A, was added at an amount of 5 % by weight and dry-blended. The resulting blend was extruded at 160 °C at 250 rpm to give a clay-dispersed poly(acrylonitrile-co-butadiene-co-styrene) copolymer nanocomposite in which the silicate of the organophilic clay was completely delaminated to silicate layers.

EXAMPLE 7

One Pot-Type Preparation of Clay-Dispersed
Poly(vinylchloride) Copolymer Nanocomposite Using
Single-Screw Extruder

5 To a polymer matrix in which poly(ϵ -caprolactone)
with a molecular weight of 10,000 was blended at an
amount of 30 % by weight with a poly(vinylchloride)
polymer, organophilic clay, such as that manufactured
10 by Southern Clay Products Inc. U.S.A., identified as
Closite 30A, was added at an amount of 5 % by weight
and dry-blended. The resulting blend was extruded at
160 °C at 250 rpm through a single-screw extruder to
give a clay-dispersed poly(vinylchloride) polymer
nanocomposite in which the silicate of the organophilic
15 clay was completely delaminated to silicate layers.

INDUSTRIAL APPLICABILITY

Over the used polymers themselves, the clay-
dispersed polymer nanocomposites according to the
present invention were measured to be improved by 30-
20 50% in elastic modulus, by 5-10% in mechanical
properties, and by 50 °C in heat deflection
temperature. With these advantages, the nanocomposites
according to the present invention can replace
conventional ABS resin and various PVC resins for use
25 in housing electronic appliances.

The present invention has been described in an
illustrative manner, and it is to be understood that

the terminology used is intended to be in the nature of description rather than of limitation. Many modifications and variations of the present invention are possible in light of the above teachings.

5 Therefore, it is to be understood that within the scope of the appended claims, the invention may be practiced otherwise than as specifically described.

CLAIMS

1. A method for the preparation of a clay-dispersed polymer nanocomposite, which comprises introducing poly(ϵ -caprolactone) , alone or in
5 combination with a thermodynamically compatible resin, as a matrix resin.

2. The method as set forth in claim 1, wherein the thermodynamically compatible resin is chlorinated polyethylene or poly(vinyl chloride).

10 3. The method as set forth in claim 1, wherein the thermodynamically compatible resin is selected from the group consisting of a poly(styrene-co-acrylonitrile)copolymer, a poly(acrylonitrile-co-butadiene-co-styrene) copolymer, and a poly(vinyl
15 chloride).

4. The method as set forth in claim 3, wherein the poly(ϵ -caprolactone) ranges, in molecular weight, from 10,000 to 100,000.

5. The method as set forth in claim 1, wherein
20 the poly(ϵ -caprolactone) is mixed with a thermodynamically compatible resin, along with an intercalant, and extruded altogether, said thermodynamically compatible resin being selected from the group consisting of a poly(styrene-co-acrylonitrile)copolymer, a poly(acrylonitrile-co-
25 acrylonitrile)copolymer, a poly(acrylonitrile-co-

butadiene-co-styrene) copolymer, and a poly(vinyl chloride).

6. The method as set forth in claim 1, wherein the poly(ϵ -caprolactone) is mixed with an organophilic
 5 clay and extruded or mixed to give a master batch, then, blended with a thermodynamically compatible resin, and extruded.

7. The method as set forth in claim 1, wherein the clay-dispersed polymer composite comprises clay
 10 which is intercalated by an intercalant.

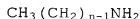
8. The method as set forth in claim 7, wherein the intercalant is selected from the group consisting of hydroxyethylmethyloctadecylammonium chloride, represented by the following chemical formula 1;

15 Chemical Formula 1



Amines, represented by the following chemical formula 2;

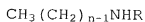
20 Chemical Formula 2



wherein n is 8, 10, 12, 16 or 18.

secondary amines represented by the following chemical formula 3:

25 Chemical Formula 3



wherein n is an integer of 8-18 and R is a hydrocarbon.

9. The method as set forth in claim 1, wherein the clay-dispersed polymer composite comprises clay which contains montmorillonite.

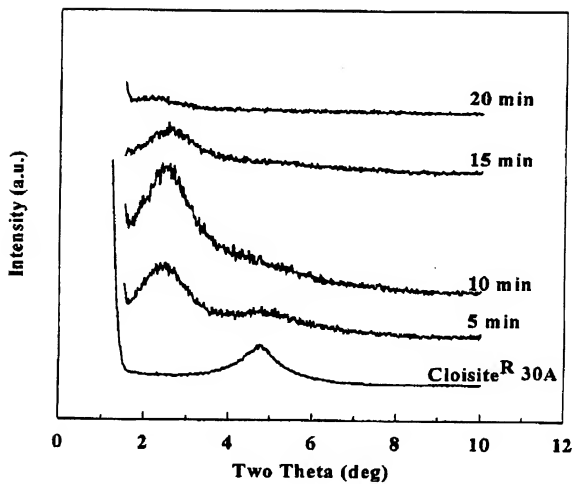
10. The method as set forth in claim 1, wherein
5 the poly(ϵ -caprolactone) is used at an amount of 20-40 % by weight based on the total weight of the nanocomposite.

ABSTRACT

Disclosed is a method for preparing a clay-dispersed polymer nanocomposite. In this method, a polymer, which carries oxygen atoms within the repeating units of its backbone and is thermodynamically compatible with a binder resin, is used as a matrix resin. Useful is poly(ϵ -caprolactone) owing to its thermodynamic compatibility with poly(styrene-co-acrylonitrile) copolymers, poly(acrylonitrile-co-butadiene-co-styrene) copolymers, and poly(vinylchloride) resins. Poly(ϵ -caprolactone) resins aid the binder resins to penetrate into silicate layers so that the silicate of the organophilic clay was completely delaminated to silicate lamellas.

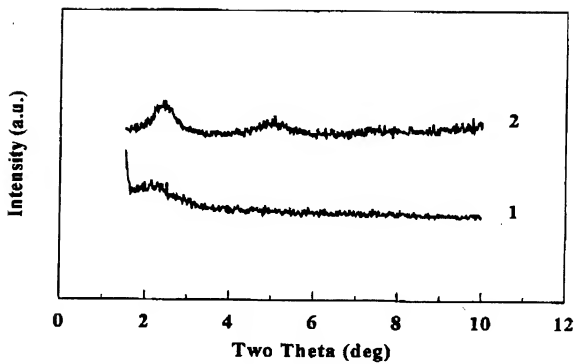
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FIG. 1a



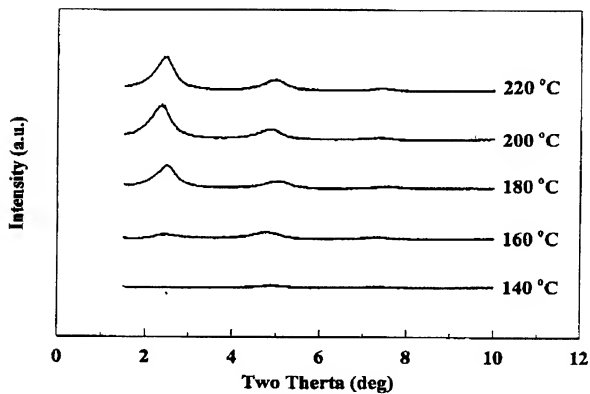
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FIG. 1b



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FIG. 2



DECLARATION FOR PATENT APPLICATION

Docket Number (Optional)
MR2685-110

As a below named inventor, I hereby declare that:

My residence, post office address and citizenship are as stated below next to my name.

I believe I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural names are listed below) of the subject matter which is claimed and for which a patent is sought on the invention entitled
METHOD FOR PREPARATION OF CLAY-DISPERSED POLYMER NANOCOMPOSITE, the specification of which is
attached hereto unless the following box is checked:

☐ was filed on _____ as United States Application Number or PCT International Application
Number _____ and was amended on _____ (If applicable).

I hereby state that I have reviewed and understand the contents of the above identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR §1.56.

I hereby claim foreign priority benefits under 35 U.S.C. § 119(a)-(d) or § 385(b) of any foreign application(s) for patent or inventor's certificate, or § 365(a) of any PCT International application which designated at least one country other than the United States, listed below and have also identified below, by checking the box, any foreign application for patent or inventor's certificate, or PCT International application having a filing date before that of the application on which priority is claimed.

Prior Foreign Application(s)

Priority Not Claimed

2000-52689	KR	6 September 2000	<input type="checkbox"/>
(Number)	(Country)	(Day/Month/Year Filed)	
_____	_____	_____	<input type="checkbox"/>
(Number)	(Country)	(Day/Month/Year Filed)	

I hereby claim the benefit under 35 U.S.C. § 119(e) of any United States provisional application(s) listed below.

(Application Number) _____ (Filing Date) _____

(Application Number) _____ (Filing Date) _____

I hereby claim the benefit under 35 U.S.C. § 120 of any United States application(s), or § 365(c) of any PCT International application designating the United States, listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States or PCT International application in the manner provided by the first paragraph of 35 U.S.C. § 112.

I acknowledge the duty to disclose information which is material to patentability as defined in 37 CFR § 1.56 which became available between the filing date of the prior application and the national or PCT International filing date of this application.

PCT/KR00/01205	24 October 2000	Pending
(Application Number)	(Filing Date)	(Status-patented, pending, abandoned)

(Application Number)	(Filing Date)	(Status-patented, pending, abandoned)
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I hereby appoint the following attorney(s) and/or agent(s) to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith:

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I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

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